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[Material]	Drawings	1
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[Name of the Document] Specification

[Title of the Invention] Stacked Photovoltaic Element

5 [Claim(s)]

[Claim 1] A stacked photovoltaic element characterized by comprising a plurality of unit photovoltaic elements each composed of a pn- or pin-junction, connected to each other in series, wherein a  
10 zinc oxide layer is provided at least one position between the unit photovoltaic elements, and the zinc oxide layer has resistivity varying in a thickness direction thereof.

[Claim 2] The stacked photovoltaic element  
15 according to Claim 1, characterized in that zinc oxide of the zinc oxide layer on a side of being in contact with a p-layer has a higher resistivity than that on a side of being in contact with an n-layer.

[Claim 3] The stacked photovoltaic element  
20 according to Claim 2, characterized in that a resistivity of the zinc oxide continuously decreases in the zinc oxide layer from a side of the zinc oxide layer in contact with the p-layer towards a side of the zinc oxide layer in contact with the n-layer.

25 [Claim 4] The stacked photovoltaic element according to any one of Claims 1 to 3, characterized in that a resistivity of zinc oxide of the zinc oxide layer

is  $2 \times 10^0 \Omega\text{cm}$  or more and  $5 \times 10^3 \Omega\text{cm}$  or less.

[Claim 5] The stacked photovoltaic element according to any one of Claims 1 to 4, characterized in that a high resistant portion of zinc oxide of the zinc  
5 oxide layer has  $5 \times 10^2 \Omega\text{cm}$  or more and  $5 \times 10^3 \Omega\text{cm}$  or less.

[Claim 6] The stacked photovoltaic element according to any one of Claims 1 to 5, characterized in that at least one of the plurality of the unit  
10 photovoltaic elements has a pin-junction comprising an i-type layer composed of amorphous Si:H.

[Claim 7] The stacked photovoltaic element according to any one of Claims 1 to 5, characterized in that at least one of the plurality of the unit  
15 photovoltaic elements has a pin-junction comprising an i-type layer composed of microcrystalline Si.

[Claim 8] The stacked photovoltaic element according to any one of Claims 1 to 5, characterized in that at least one of the plurality of the unit  
20 photovoltaic elements has a pin-junction comprising an i-type layer composed of single-crystalline or polycrystalline Si.

[Detailed Description of the Invention]

[0001]

25 [Field of the Invention]

The present invention relates to a stacked photovoltaic element comprising at least two power-

generating function units.

[0002]

[Prior Art]

A photovoltaic element is a device which converts  
5 incident light energy into electric energy. A solar  
cell is a photovoltaic element which converts solar  
rays as white light into electric energy. It is  
characterized by efficiently converting light over a  
wide wavelength range into electric energy. It is  
10 necessary to efficiently absorb light over a wide  
wavelength range in order to achieve a high conversion  
efficiency. A stacked photovoltaic element, which is  
formed by stacking having a semiconductor layer having  
a different band gap from each other as the photoactive  
15 layer is known as a solution. This stacked  
photovoltaic element has one element having a  
semiconductor of relatively larger band gap on a light  
incident side, and another element having a  
semiconductor of relatively smaller band gap in this  
20 order from the light incident side, the former  
absorbing light of shorter wavelengths with large  
energy and the latter absorbing light of longer  
wavelengths with small energy which the former  
transmits. The stacked photovoltaic element,  
25 therefore, can more efficiently absorb and utilize  
light over a wider wavelength range.

[0003]

It is essential to provide each photovoltaic element with light of a wavelength in a range suitable for that element, because a wavelength range of incident light which each photovoltaic element can  
5 utilize is restricted depending on the band gap of a semiconductor used as the photoactive layer for that element. In other words, photon cannot be absorbed by the semiconductor when photon has lower energy than the band gap of the semiconductor. In such a case, it only  
10 passes through the semiconductor without being utilized. On the other hand, photon having higher energy than the band gap of the semiconductor cannot be fully utilized although it can be absorbed, because potential energy of electron which can be produced when  
15 the electron is excited is limited by magnitude of the band gap, whereby the difference between the band gap energy and photon energy cannot be utilized. In other words, it is essential to design a stacked photovoltaic element to selectively introduce light of shorter  
20 wavelength range into a photovoltaic element on the incident light side and light of longer wavelength range into another photovoltaic element arranged under the above element.

[0004]

25 One of the known solutions is to provide a transparent electroconductive film as a light-reflecting layer between these photovoltaic elements.



For example, Patent Document 1 or Non-patent Document 1 disclose a method for providing an electroconductive intermediate layer each between the elements which reflects light of shorter wavelength and transmit light of longer wavelength. Patent Document 2 discloses a method for adjusting the thickness of a selective reflection layer in such a way to increase electric current flowing through a photovoltaic element on the incident light side by setting its peak reflectivity to the maximum wavelength of spectral sensitivity of the photovoltaic element on the incident light side. Patent Document 3 discloses a method for enhancing efficiency of a stacked photovoltaic element by a selective reflection layer of stacked structure having a higher reflectivity for the shorter wavelength range which the upper photoelectron conversion layer can absorb more easily, and a lower reflectivity for the longer wavelength range which the lower photoelectron conversion layer can absorb more easily for transmission. Each of these techniques uses a dielectric layer of SnO<sub>2</sub>, ZnO, ITO or the like as the selective reflection layer, to prevent light of short wavelength, which should be absorbed by the photovoltaic element on the incident light side, from being absorbed by the lower photovoltaic element and thereby to enhance conversion efficiency of the photovoltaic element on the incident light side.

[0005]

[Patent Document 1]

Japanese Patent Application Laid-Open No. S63-  
77167

5 [Patent Document 2]

Japanese Patent Application Laid-Open No. H2-  
237172

[Patent Document 3]

Japanese Patent Application Laid-Open No. 2001-  
10 308354

[Non-patent Document 1]

Kenji Yamamoto, "Thin-film polycrystalline silicon  
solar cell", Applied Physics, The Japan Society of  
Applied Physics, May, 2002, Vol.71, No.5, p.524 to 527

15 [0006]

[Problem to be Solved by the Invention]

On the other hand, the following problems occur  
when the above-described electroconductive reflection  
layer is provided as the intermediate layer.

20 [0007]

A photovoltaic element of large area, comprising  
unit elements stacked on each other in series, e.g., as  
shown in Fig. 1, has electrical defects within the  
element, resulting from dust generated during the  
25 deposition step, or irregularities or foreign matter on  
the surface of a substrate. In Fig. 1, numeral 100  
denotes the stacked photovoltaic element; 101:

substrate, 102: second photovoltaic element, 103: zinc oxide layer, 104: first photovoltaic element, 105: electroconductive layer as transparent electrode, 106: short circuit in the second photovoltaic element, and  
5 107: short circuit in the first photovoltaic element. The electrical defects inevitably associated with the large area deteriorate the element characteristics resulting from decreased shunt resistance and fill factor (FF). One of the effective means to solve these  
10 problems is a method (passivation) in which a photovoltaic element is normally dipped in an electrolytic solution and an electric current is passed through it to selectively remove a part of an electroconductive layer outside of an electric defect.  
15 However, for a stacked photovoltaic element comprising a lower photovoltaic element layer, an intermediate layer, an upper photovoltaic element layer and an electroconductive layer on a substrate, the above procedure can partly remove the electroconductive layer  
20 105 on the first photovoltaic element layer as the upper layer containing the defect 107, but cannot remove the intermediate layer 103 on the second photovoltaic element layer as the lower layer containing the defect 106. As a result, short-circuit  
25 current flows through the defect in the lower layer to decrease electromotive force of the lower photovoltaic element. Short-circuit current cannot be effectively

prevented from spreading into the intermediate layer for various reasons. In particular the intermediate layer should have a certain thickness to function as the reflection layer, and should satisfy the other  
5 considerations, e.g., compatibility with the semiconductor layers with which it is in contact on both sides and series resistance. These requirements limit a range in which the resistivity of the materials can be adjusted. Moreover, additional junctions  
10 generated between the photovoltaic elements and an intermediate layer of a different material therebetween inevitably deteriorate characteristics which are accompanied by decreased FF. Still more, a plurality of layers inserted to prevent a reduced shunt  
15 resistance further increase junction number, thus further aggravating the interfacial problems.

[0008]

As discussed above, incorporation of an intermediate layer as a selective reflection layer for  
20 increasing photocurrent involves the adverse effect of decreased electromotive force of the photovoltaic element.

[0009]

The present invention is to provide a photovoltaic  
25 element exhibiting a high conversion efficiency realized by producing large photocurrent without causing decreased electromotive force.

[0010]

[Means for Solving the Problem]

In order to achieve the object mentioned above, a stacked photovoltaic element according to the present invention is characterized by comprising a plurality of unit photovoltaic elements each composed of a pn- or pin-junction, connected to each other in series, wherein at least a zinc oxide layer is provided at least one position between the unit photovoltaic elements, and the zinc oxide layer has resistivity varying in the thickness direction.

[0011]

Zinc oxide on a side of being in contact with a p-layer of the pn- or pin-junction has a higher resistivity than that on a side of being in contact with an n-layer of the pn- or pin-junction in the zinc oxide layer.

[0012]

The resistivity of zinc oxide continuously decreases from the p-layer side towards the n-layer side.

[0013]

The resistivity of zinc oxide is preferably  $2 \times 10^0$   $\Omega\text{cm}$  or more and  $5 \times 10^3$   $\Omega\text{cm}$  or less.

[0014]

The highest resistant portion of zinc oxide preferably has  $5 \times 10^2$   $\Omega\text{cm}$  or more and  $5 \times 10^3$   $\Omega\text{cm}$  or less.

[0015]

Of the plurality of the unit photovoltaic elements for the first aspect, at least one unit photovoltaic element has a pin-junction comprising an i-type layer  
5 suitably composed of an amorphous Si:H.

[0016]

Of the plurality of the unit photovoltaic elements for the first aspect, at least one unit photovoltaic element has a pin-junction comprising an i-type layer  
10 suitably composed of microcrystalline Si.

[0017]

Of the plurality of the unit photovoltaic elements for the first aspect, at least one unit photovoltaic element has a pin-junction comprising an i-type layer  
15 suitably composed of single-crystalline or polycrystalline Si.

[0018]

[Embodiment(s)]

Fig. 3 outlines a cross-sectional structure of the  
20 two-layers-stacked photovoltaic element 300 as one embodiment of the present invention. It has on a substrate 301 of a metal or the like which has a reflection layer stacked thereon, the second photovoltaic element 302, second zinc oxide layer 303,  
25 first zinc oxide layer 304, first photovoltaic element 305 and transparent electrode 306 stacked in this order. A semiconductor which constitutes the

photoactive section for the first photovoltaic element 305 has a larger band gap than that of a semiconductor which constitutes the photoactive section for the second photovoltaic element 302. This means that the stacked photovoltaic element 300 is designed in such a way that the first photovoltaic element 305 absorbs light in a short wavelength range while the second photovoltaic element 302 absorbs light in a long wavelength range. The first zinc oxide layer 304 and first photovoltaic element 305 have a different refractive index. Therefore, multiple reflection can be created by adjusting the thickness of each layer to efficiently enhance reflectivity in the short wavelength range, and thereby to increase the quantity of light absorbed by the first photovoltaic element 305. The second zinc oxide layer 303 is designed to have a higher resistivity than that of the first zinc oxide layer 304.

[0019]

Fig. 4 schematically illustrates power-generating operation of the stacked photovoltaic element of the present invention. An electrical defect in the first photovoltaic element acts as a short-circuit current passage. When the electrical defect 402 in the first photovoltaic element 305 is close to the electrical defect 403 in the second photovoltaic element 302, no deterioration of the photovoltaic element

characteristics is caused by these defects, because the transparent electrode 306 is removed by the shunt passivation treatment carried out after the stacked photovoltaic element is assembled. When these defects  
5 are apart from each other at a distance, on the other hand, the characteristics may be deteriorated, because of insufficient shunt passivation treatment. Although the first photovoltaic element 305 is provided with the first zinc oxide layer 304 of lower resistivity on the  
10 substrate side surface, it is sufficiently thin and causes spread of short-circuit current in the lateral direction to only a limited extent. Therefore, it little deteriorates the characteristics.

[0020]

15 On the other hand, in the vicinity of the electrical defect in the second photovoltaic element 302, the electroconductive zinc oxide layer is present, and the defect acts as a short circuit in the photovoltaic element while power is being generated.  
20 However, the second photovoltaic element 302 has the second zinc oxide layer 303 of higher resistivity on the surface of the element 302 (upper layer surface), whereby short-circuit current spreads via the first zinc oxide layer 304 of lower resistivity while its  
25 spread in the lateral direction is limited. A total thickness of the first zinc oxide layer 304 and second zinc oxide layer 303 is limited to a level suitable for



the selective reflection layer, and spread of the short-circuit current can be efficiently prevented by combination of the higher and lower resistivities.  
[0021]

5           The electrical polarities at each junction of the stacked structure consisting of the second photovoltaic element, second and first zinc oxide layer and first photovoltaic element provided in this order are, for example, as shown in Fig. 2, a combination of n-  
10   /n+/n++, i.e., the second zinc oxide layer 202 of the lowest carrier concentration (n-), the first zinc oxide layer 203 of higher carrier concentration (n+) and the n-type semiconductor layer 204 of the first photovoltaic element of the highest carrier  
15   concentration (n++) are stacked in this order. In Fig. 2, 201 denotes a substrate, 202: second zinc oxide layer of n- type, 203: first zinc oxide layer of n+ type, 204: first photovoltaic element as the n-type semiconductor layer of n++ type, 205: transparent  
20   electrode, 206: second photovoltaic element, 207: stacked zinc oxide layer, and 208: first photovoltaic element.

[0022]

25           The n-type zinc oxide layer can have a controlled bulk carrier concentration by the production conditions. It is considered that light-generating carrier can be collected more efficiently by increasing

carrier concentration in the n-type zinc oxide layer  
stepwise or continuously from the p-type semiconductor  
layer side of the second photovoltaic element towards  
the n-type semiconductor layer side of the first  
5 photovoltaic element, because increasing or decreasing  
trend of concentration of the carrier of the same  
polarity is set in one direction including the n-type  
semiconductor layer of the first photovoltaic element,  
thereby efficiently connecting these elements to each  
10 other at the band junction, although the concept is not  
fully substantiated.

[0023]

The zinc oxide layer as the intermediate layer has  
a higher resistivity, preferably  $2 \times 10^0 \Omega\text{cm}$  or more and  
15  $5 \times 10^3 \Omega\text{cm}$  or less for both the first and second zinc  
oxide layers, than recent zinc oxide layers of which  
the resistivity has been greatly decreased. The  
intermediate layer having a resistivity in the above  
range has a lower carrier concentration than that of  
20 the n-type semiconductor layer of the first  
photovoltaic element, conceivably resulting in improved  
junction. On the other hand, short-circuit current is  
considered to diffuse in the zinc oxide layer, unless  
zinc oxide has a resistivity of  $5 \times 10^2 \Omega\text{cm}$  or more and  
25  $5 \times 10^3 \Omega\text{cm}$  or less in the high-resistivity portion, to  
deteriorate the characteristics.

[0024]

The zinc oxide layer preferably transmits at least 50% of light having a wavelength of 800 nm. The wavelength range of solar light which can be effectively utilized by the photovoltaic element is  
5 approximately from 300 to 1200 nm in consideration of its spectral pattern. The cell at the upper section of the zinc oxide layer absorbs light of short wavelength, and hence it is preferable for the zinc oxide layer to effectively transmit light of long wavelength.  
10 Therefore, it preferably transmits at least 50% of light having a wavelength of 800 nm, which represents the long wavelength.

[0025]

Hereinafter, the embodiments describe example for  
15 a solar cell as a stacked photovoltaic element comprising two photovoltaic layers (unit photovoltaic elements), produced by stacking on a substrate, a reflection layer, a photovoltaic element of microcrystalline silicon (hereinafter referred to as  
20 the "bottom cell"), an intermediate layer of zinc oxide and a photovoltaic element of amorphous silicon (hereinafter referred to as the "top cell") in this order from the substrate side. However, the present invention is by no means limited to the above  
25 structure, and the number of the photovoltaic elements may be increased as required.

[0026]

Next, each component for the present invention is described in detail.

[0027]

[Substrate]

5           The material for the substrate which constitutes the stacked photovoltaic element of the present invention is not limited. It may be an electroconductive or insulating material of any type. The electroconductive materials include metals, e.g.,  
10   plated steel, NiCr, stainless steel, Al, Cr, Mo, Au, Nb, Ta, V, Ti, Pt, Pb, Sn and an alloy thereof. The insulating materials include synthetic resins, e.g., polyester, polyethylene, polycarbonate, cellulose acetate, polypropylene, polyvinyl chloride,  
15   polyvinylidene chloride, polystyrene and polyamide; and glass, ceramics and paper. The particularly preferable materials are stainless steel as the metal substrate, and glass, ceramics and polyimide as the insulating substrate. A transparent, insulating substrate is used  
20   for the stacked photovoltaic element with incident light entering from the substrate side, and glass is suitable for the substrate.

[0028]

25           The substrate surface may be smooth or of irregular texture with irregularities having the maximum height of 0.1 to 1.0  $\mu\text{m}$ . A stainless steel substrate may have a surface of irregular texture by

etching with an acidic solution.

[0029]

Thickness of the substrate may be adequately set to give a desired stacked photovoltaic element by  
5 stacking the layers one on another as intended. When the stacked photovoltaic element is required to be flexible, it may be as thin as possible so long as it can sufficiently work as a support. However, it normally has a thickness of 10  $\mu\text{m}$  or more in  
10 consideration of production, handling and mechanical strength.

[0030]

[Reflection Layer]

The reflection layer for the stacked photovoltaic  
15 element of the present invention comprises of a metal reflection layer and an increased reflection layer. The metal reflection layer is made of a deposited film of a metal which has a high reflectivity of light from the visible to near-infrared ray, e.g., Ag, Al, Cu and  
20 an alloy thereof. It is suitably formed by, e.g., vacuum evaporation, sputtering or electrolytic deposition from an aqueous solution. It is preferably 10 to 5000 nm thick. It is preferably provided with irregularities on the surface for irregular reflection.  
25 It is also preferably provided with an increased reflectance layer to increase quantity of reflected light.

[0031]

The materials for the increased reflectance layer include ZnO, SnO<sub>2</sub>, In<sub>2</sub>O<sub>3</sub>, ITO, TiO<sub>2</sub>, CdO, Cd<sub>2</sub>SnO<sub>4</sub>, Bi<sub>2</sub>O<sub>3</sub>, MoO<sub>3</sub> and Na<sub>x</sub>WO<sub>3</sub>. The increased reflectance layer is  
5 suitably formed by vacuum evaporation, sputtering, electrolytic deposition, CVD, spraying, spin-on, dipping or the like of the above material. Its thickness is preferably 50 nm to 10 μm, although the optimum thickness varies depending on inherent  
10 reflectivity of the material used. The increased reflectance layer is preferably provided with irregularities on the surface for increasing light scattering. For example, in the sputtering method sputtering conditions may be selected to provide  
15 irregularities based on the grain boundaries.

[0032]

[Photovoltaic Layer]

The semiconductors useful for the stacked photovoltaic element of the present invention include  
20 single-crystalline, poly-crystalline, microcrystalline and amorphous materials of IV, III-V, II-VI, I-III-VI<sub>2</sub> groups. The IV group materials include C, Si and Ge and an alloy thereof, III-V group materials AlAs, AlSb, GaN, GaP, GaAs, GaSb, InP and InAs, II-VI group  
25 materials ZnSe, ZnS, ZnTe, CdS, CdSe, CdTe and Cu<sub>2</sub>S, and I-III-VI<sub>2</sub> group materials CuInSe<sub>2</sub>. Of these, silicon-based semiconductors are more preferable. The

single-crystalline, poly-crystalline, microcrystalline  
and amorphous semiconductors are suitably used.

[0033]

The photovoltaic layer for the stacked  
5 photovoltaic element of the present invention contains  
a pn- or pin-junction.

[0034]

The stacked photovoltaic element of the present  
invention comprises at least two photovoltaic layers.  
10 The semiconductors for these photovoltaic layers may be  
the same or different material. However, in the  
preferable structure, a photovoltaic layer using a  
semiconductor which can more efficiently absorb light  
of shorter wavelengths and another photovoltaic layer  
15 using a semiconductor which can more efficiently absorb  
light of longer wavelengths are stacked in this order  
from the incident light side, because light of shorter  
wavelength is more easily absorbed.

[0035]

20 [Zinc Oxide Layer]

The stacked photovoltaic element of the present  
invention has an intermediate layer composed of zinc  
oxide in at least one position between the photovoltaic  
layers (unit photovoltaic elements).

25 [0036]

The zinc oxide layer for the present invention can  
be suitably formed by vacuum evaporation, DC magnetron

sputtering, RF magnetron sputtering, electrolytic deposition, electroless plating, CVD, MOCVD, spraying, spin-on, dipping, sol-gel process or the like. The generally known dopants for adjusting its resistivity  
5 include Al, B, Ga and In. The other known dopants include tetravalent metals, e.g., Si, Ge, Ti and Zr. When a common vacuum evaporation or sputtering method is employed, zinc oxide (target or the like) may be sintered after previously adding the dopant at a  
10 desired content to zinc oxide.

[0037]

The zinc oxide layer has a varying reflectivity to reflect more light having a wavelength shorter than the wavelength  $\lambda_m$ , at which the second photovoltaic element  
15 attains the highest spectral characteristics, and to reflect less light having a wavelength longer than the wavelength  $\lambda_m$ , in order to efficiently convert energy of incident light over an entire wavelength range. The zinc oxide layer preferably has a transmittance of 80%  
20 or more, to efficiently utilize incident light.

[0038]

The zinc oxide layer for the present invention has a resistivity varying in the thickness direction to prevent deteriorated element characteristics caused by  
25 shunt, as the problem involved in the conventional layer, while keeping its function as a selective reflection film. Its resistivity is preferably  $2 \times 10^0$



$\Omega\text{cm}$  or more and  $5 \times 10^3 \Omega\text{cm}$  or less. The high resistivity portion of the zinc oxide layer is preferably  $5 \times 10^2 \Omega\text{cm}$  or more and  $5 \times 10^3 \Omega\text{cm}$  or less. Its thickness is preferably 0.2 to  $2\mu\text{m}$  in consideration  
5 of its reflectivity, series resistance and irregularities on the surface.

[0039]

[Transparent Electrode]

The materials for the transparent electrode for  
10 the stacked photovoltaic element of the present invention include indium oxide, tin oxide, indium-tin oxide or zinc oxide. The transparent electrode is formed by sputtering, vacuum evaporation, chemical vapor deposition, ion plating, ion beam or ion beam  
15 sputtering. It may be also formed by electrolytic deposition or immersion in an aqueous solution of a metallic ion, e.g., that containing nitric, acetic or ammonia group. It preferably has a sufficient thickness to satisfy the requirements as an  
20 antireflection film.

[0040]

[Examples]

The examples as the preferred embodiments of the present invention are described in detail by referring  
25 to the attached drawings, however, the present invention is not limited to these embodiments.

[0041]

[Example 1]

In Example 1, the stacked photovoltaic element having the pin-type second photovoltaic element 302 with the i-layer of intrinsic microcrystalline Si, pin-type first photovoltaic element 305 with the i-layer of intrinsic amorphous Si:H and the intermediate layer of zinc oxide (refer to Fig. 3) was produced.

[0042]

The substrate 301, 45 mm square and 0.15 mm thick, was of flat stainless steel (SUS 430), commonly referred to as BA-finished one. It was put in a commercial DC magnetron sputtering unit (not shown), which was evacuated to a pressure of  $10^{-3}$  Pa or less.

[0043]

Argon was blown into the unit at  $30 \text{ cm}^3/\text{min}$  (normal conditions) to keep pressure inside at  $2 \times 10^{-1}$  Pa. A DC power of 120 W was applied to an aluminum target (diameter: 6 inches) for 90 seconds to form a thin film of aluminum with a thickness of 70 nm on the substrate, while the substrate was kept unheated. Then, a DC power of 500 W was applied to a zinc oxide target (diameter: 6 inches), after the electrical connection was changed, for 30 minutes to form the reflection layer of zinc oxide with a thickness of about 500 nm, while the substrate was heated at  $200^\circ\text{C}$ .

[0044]

Fig. 5 schematically illustrates one embodiment of

suitable apparatus for producing a semiconductor layer for the stacked photovoltaic element of the present invention, where the deposited film forming apparatus 500 comprises the following major components; 501: load  
5 chamber, 503: chamber for the i-type layer of microcrystalline silicon, 504: RF chamber for the i-type layer of amorphous silicon, 502: RF chamber for the n-type layer, 505: RF chamber for the p-type layer and 506: unload chamber. These chambers are isolated  
10 from each other by a gate valve 507, 508, 509, 510 or 511 so that feed gases of the respective chambers are not mixed with each other.

[0045]

The chamber 503 for the i-type layer of  
15 microcrystalline silicon is composed of the heater 512 for heating the substrate and plasma CVD chamber 513. The RF chamber 502 is composed of the heater 514 for depositing the n-type layer and deposition chamber 515 for depositing the n-type layer, the RF chamber 504 the  
20 heater 516 for depositing the i-type layer and deposition chamber 517 for depositing the i-type layer, and the RF chamber 505 the heater 518 for depositing the p-type layer and deposition chamber 519 for depositing the p-type layer. The substrate is  
25 supported by the substrate holder 521 to run on the rail 520 by a roller driven from an outside power source. In the plasma CVD chamber 513, a microcrystal

is deposited by microwave plasma CVD or VHF plasma CVD.  
[0046]

The deposited film forming apparatus of the above structure was used to form, as the second photovoltaic  
5 element 302, pin-type photovoltaic element whose i-type layer was of intrinsic microcrystalline Si under the deposition conditions given in Table 1 by the following procedure.

[0047]

10 First, the substrate 301 provided with a reflection layer was set on the substrate holder 521 and then on the rail 520 in the load chamber 501. The load chamber 501 was then evacuated to a vacuum of several hundreds mPa or less.

15 [0048]

Next, the gate valve 507 was opened, and the substrate holder 521 was moved into the deposition chamber 515 for depositing the n-type layer in the chamber 502, where the n-type layer was deposited to a  
20 given thickness using a given feed gas, while the gate valves 507, 508, 509, 510 and 511 were kept closed. The chamber was sufficiently evacuated, and the gate valve 508 was opened to move the substrate holder 521 into the deposition chamber 503. Then the gate valve  
25 508 was closed.

[0049]

The substrate was heated to a given temperature by

the heater 512, a necessary quantity of the feed gas was charged in the chamber, and given microwave or VHF energy was introduced into the deposition chamber 513 which was evacuated to a given vacuum level, to  
5 generate a plasma therein to deposit the i-type layer of microcrystalline silicon to a given thickness. The chamber 503 was sufficiently evacuated, and the gate valves 509 and 510 were opened to move the substrate holder 521 from the chamber 503 to the chamber 505.  
10 [0050]

After the substrate holder 521 was moved into the deposition chamber 519 for depositing the p-type layer in the chamber 505, the substrate was heated to a given temperature by the heater 518. A necessary quantity of  
15 the feed gas to deposit the p-type layer was charged into the deposition chamber 519 into which RF energy was introduced to deposit the p-type layer to a given thickness, while the chamber was kept at a given vacuum level.

20 [0051]

Similarly as in the above, the deposition chamber 519 was sufficiently evacuated, and the gate valve 511 was opened to move the substrate holder 521 into the unload chamber 506. Then, all gate valves were closed  
25 and nitrogen gas was charged into the unload chamber 506, and the substrate was cooled with nitrogen gas in the unload chamber 506, while all of the gate valves

were closed. Then, the substrate holder 521 was taken out of the unload chamber 506, after the discharge valve was opened.

[0052]

[Table 1]

		Gas for layer formation (cm <sup>3</sup> /minute under normal conditions)				Power density (W/cm <sup>2</sup> )		Pressure (Pa)	Substrate temperature(°C)	Thickness of layer (nm)
		SiH <sub>4</sub>	H <sub>2</sub>	PH <sub>3</sub> (diluted to 2% with H <sub>2</sub> )	BF <sub>3</sub> (diluted to 2% with H <sub>2</sub> )	RF	VHF			
Second photovoltaic element	N2	2	48	0.5		0.04		180	225	20
	I2	25	750				0.2	40	250	2000
	P2	0.025	35		1	1.2		270	165	5

[0053]

Next, the substrate, on which the layers up to the second photovoltaic element 302 was formed, was removed from the substrate holder 521, and set on the substrate holder 601 in the DC magnetron sputtering apparatus 600 for forming a zinc oxide layer, shown in Fig. 6. Then, the apparatus was evacuated to a pressure of  $10^{-3}$  Pa or less.

[0054]

10       The substrate holder 601 was electrically insulated, and could keep the photovoltaic element as the sample floated. The apparatus was kept at a pressure of  $2 \times 10^{-2}$  Pa while argon gas was supplied at 50 sccm, and oxygen gas and vaporized  $H_2O$  gas were  
15       supplied at a rate given in Table 2 within 0.1 to 5 sccm, all via the gas inlet tube 602. The substrate holder 601 was heated by the heater 603 to keep the substrate at  $150^\circ\text{C}$ . A DC power of 500 W was applied from the DC power source 605 onto the Al-doped zinc  
20       oxide (ZnO) target 604 (diameter: 6 inches) for 10 minutes, to deposit the second zinc oxide layer 303 with a thickness of about  $0.5 \mu\text{m}$  on the substrate. The target 604 was surrounded by the earth shield 606, to prevent diffusion of the plasma and to stabilize the  
25       discharge. At the same time, a quartz substrate (45 by 45 mm) was set on the substrate holder to deposit thereon the same layer of zinc oxide for the analysis



of its electrical properties.

[0055]

Next, on each of the zinc oxide layers 303, Samples A to J given in Table 2 formed on each  
5 substrate, the first zinc oxide layer 304 was deposited under the conditions of A to J as given in Table 2. A total of 100 samples were prepared. At the same time, a quartz substrate (45 by 45 mm) was set on the  
substrate holder to deposit thereon the same layer of  
10 zinc oxide for the analysis of its electrical properties.

[0056]

[Table 2]

Sample No.	Oxygen flow rate (sccm)	H <sub>2</sub> O flow rate (sccm)
A	0.1	10
B	0.3	10
C	0.4	10
D	0.5	10
E	0.8	10
F	1.0	10
G	2.0	5
H	5.0	5
I	10.0	2
J	15.0	2

[0057]

15 Next, the deposited film forming apparatus 500 was

again used to form, on a substrate on which the above intermediate layer (zinc oxide layer) was formed, as the first photovoltaic element 305, pin-type photovoltaic element whose i-type layer was of  
5 intrinsic amorphous Si:H under the deposition conditions given in Table 3 by the following procedure.  
[0058]

First, the n-type layer was deposited under given conditions to a given thickness in a manner similar to  
10 that described above. The gate valves 508 and 509 were opened and the substrate holder 521 was moved into the deposition chamber 504, after it was sufficiently evacuated. Then, these valves were closed.  
[0059]

15 The substrate was heated to a given temperature by the heater 516, and a necessary quantity of the feed gas was charged in the chamber. Given RF energy was introduced into the deposition chamber 517, which was evacuated to a given vacuum level, to generate a plasma  
20 therein to deposit the i-type layer of amorphous Si:H to a given thickness by adjusting deposition time. The chamber 504 was sufficiently evacuated, and the gate valve 510 was opened to move the substrate holder 521 from the chamber 504 to the chamber 505.  
25 [0060]

The p-type layer was deposited to a given thickness in a manner similar to that described above.

[0061]

Then, the deposition chamber 519 was sufficiently evacuated, and the gate valve 511 was opened to move the substrate holder 521, which carried the substrate  
5 provided with the layers up to the first photovoltaic element 305, into the unload chamber 506 also in a manner similar to that described above.

[0062]

Then, the substrate holder 521 was taken out of  
10 the unload chamber 506 also in a manner similar to that described above.

[0063]

[Table 3]

		Gas for layer formation (cm <sup>3</sup> /minute under normal conditions)				Power density (W/cm <sup>2</sup> )		Pressure (Pa)	Substrate temperature(°C)	Thickness of layer (nm)
		SiH <sub>4</sub>	H <sub>2</sub>	PH <sub>3</sub> (diluted to 2% with H <sub>2</sub> )	BF <sub>3</sub> (diluted to 2% with H <sub>2</sub> )	RF	VHF			
First photovoltaic element	N2	2	48	0.5		0.04		180	225	10
	I2	2	48			0.04		150	210	500
	P2	0.025	35		1	1.2		270	165	5

[0064]

Next, the substrate provided with the layers up to the first photovoltaic element 305 was set on the anode surface of a DC magnetron sputtering apparatus, and  
5 masked with stainless steel. Indium/tin oxide was sputtered as the transparent electrode onto the substrate in a center area of a 40 by 40 mm using a target of mixed oxide of tin (10% by weight) and indium (90% by weight).

10 [0065]

The oxide film was deposited to a 70 nm under the conditions of substrate temperature: 170°C, argon (as an inert gas) flow rate: sccm, oxygen gas flow rate: 0.5 sccm, pressure in the deposition chamber: 300 mPa,  
15 power density on the unit target area: 0.2 W/cm<sup>2</sup> and deposition time: about 100 seconds. Thickness of the film was estimated by the predetermined relationship between thickness and deposition time. The photovoltaic element sample thus prepared was named  
20 "Ex. 1".

[0066]

[Comparative Example 1]

A photovoltaic element was prepared in the same manner as in Example 1, except that no zinc oxide layer  
25 was placed between the first and second photovoltaic element.

[0067]

(Measurement)

First, the electrical characteristics of the zinc oxide layer deposited on the quartz substrate in Example 1 were measured. Resistivity of zinc oxide varies over a wide range (10 digits), and should be evaluated by an analysis system suitable for the resistance varying values. The 2-terminal method generally used for insulators is sensitive to contact resistance, and a resistivity meter (MCP-T600 manufactured by Dian Instruments company) based on the 4-terminal method was used, where a constant current was applied by the system including 4 terminals and 4 probes connected in series to measure a potential across and thereby resistivity. The results are given in Table 4. A zinc oxide layer prepared with the target containing a higher content of dopant and smaller quantities of charged  $O_2$  and  $H_2O$  had a lower resistivity.

[0068]

[Table 4]

Sample No.	Resistivity ( $\Omega\text{cm}$ )
A	$5.0 \times 10^{-1}$
B	$3.0 \times 10^0$
C	$5.5 \times 10^0$
D	$1.5 \times 10^1$
E	$5.5 \times 10^2$
F	$8.5 \times 10^2$
G	$1.4 \times 10^3$
H	$5.2 \times 10^3$
I	$8.0 \times 10^3$
J	$1.0 \times 10^4$

[0069]

Next, a total of 101 photovoltaic element samples prepared in each of Example 1 and Comparative Example 1 were measured for the current-voltage characteristics while they were irradiated with light under the conditions of AM1.5 spectral pattern and intensity of 100 mW/cm<sup>2</sup> using an analyzer (YSS-150 manufactured by Yamashita Denso company), and their short-circuit current density Jsc (mA/cm<sup>2</sup>), open voltage Voc (V) and fill factor FF were estimated based on these characteristics to determine conversion efficiency  $\eta(\%)$ .

[0070]

Shunt resistance (Rsh) and series resistance (Rs) were also estimated based on the current-voltage

characteristics under a dark condition, where the former was defined as slope near  $V=0$ , and the latter as slope of current when it rose up.

[0071]

5           The results are given in Tables 5, 6, 7 and 8, respectively.

          The zinc oxide layer having a higher resistivity on the second photovoltaic element side and a lower resistivity on the first photovoltaic element side can  
10 improve FF level, increase photocurrent and improve conversion efficiency for the stacked photovoltaic element by increasing reflection, reducing short-circuit current and improving the joint surface. On the other hand, the layer having a reversed combination  
15 of resistivity deteriorates in conversion efficiency as the  $J_{sc}$  value decreases. Moreover, the layer having the former resistivity combination exhibits excellent characteristics, when its resistivity is in a range from  $2 \times 10^0 \Omega\text{cm}$  to  $5 \times 10^3 \Omega\text{cm}$ . However, it deteriorates  
20 in conversion efficiency when its resistivity is below  $2 \times 10^0 \Omega\text{cm}$  or the high resistivity portion has  $5 \times 10^2 \Omega\text{cm}$  or less, resulting in decreased shunt resistance by short-circuit current, which in turn causes decreased FF value and then  $J_{sc}$  level. Resistivity exceeding  
25  $5 \times 10^3 \Omega\text{cm}$ , on the other hand, slightly reduces the conversion efficiency, resulting from increased series resistance.



[0072]

[Table 5]

Conversion efficiency		First zinc oxide layer									
		A	B	C	D	E	F	G	H	I	J
Second zinc oxide layer	Comparative Example 1	0.943									
	A	0.971	0.969	0.972	0.973	0.971	0.981	0.978	0.977	0.973	0.972
	B	0.970	0.972	0.975	0.972	0.976	0.980	0.981	0.982	0.981	0.978
	C	0.985	0.981	0.982	0.983	0.981	0.981	0.982	0.982	0.982	0.980
	D	0.973	0.980	0.983	0.983	0.984	0.985	0.985	0.986	0.986	0.985
	E	0.974	0.982	1.105	1.053	1.002	0.988	0.978	0.980	0.990	0.986
	F	0.982	0.990	1.101	1.110	1.058	1.015	0.991	0.992	0.990	0.990
	G	0.985	0.992	1.102	1.102	1.169	1.140	1.011	0.987	0.991	0.996
	H	0.983	0.993	1.103	1.110	1.089	1.088	1.090	1.005	0.958	0.980
	I	0.988	0.990	0.995	0.996	0.998	0.992	0.994	0.993	0.991	0.990
	J	0.986	0.987	0.991	0.992	0.995	0.991	0.993	0.991	0.990	0.989

[0073]

[Table 6]

JSC		First zinc oxide layer									
		A	B	C	D	E	F	G	H	I	J
Second zinc oxide layer	Comparative Example 1	0.960									
	A	0.971	0.969	0.972	0.973	0.971	0.981	0.978	0.977	0.973	0.972
	B	0.970	0.972	0.975	0.972	0.976	0.980	0.981	0.982	0.981	0.978
	C	0.985	0.981	0.982	0.983	0.981	0.981	0.982	0.982	0.982	0.980
	D	0.973	0.980	0.983	0.983	0.984	0.985	0.985	0.986	0.986	0.985
	E	0.974	0.982	1.105	1.053	1.002	0.988	0.978	0.980	0.990	0.986
	F	0.982	0.990	1.101	1.110	1.058	1.015	0.991	0.992	0.990	0.990
	G	0.985	0.992	1.102	1.102	1.169	1.140	1.011	0.987	0.991	0.996
	H	0.983	0.993	1.103	1.110	1.089	1.088	1.090	1.005	0.958	0.980
	I	0.988	0.990	0.995	0.996	0.998	0.992	0.994	0.993	0.991	0.990
	J	0.986	0.987	0.991	0.992	0.995	0.991	0.993	0.991	0.990	0.989

[0074]

[Table 7]

Shunt resistance		First zinc oxide layer									
		A	B	C	D	E	F	G	H	I	J
Second zinc oxide layer	A	0.956	0.955	0.960	0.954	0.946	0.952	0.953	0.954	0.956	0.963
	B	0.963	0.972	0.974	0.978	0.985	0.989	0.988	0.987	0.986	0.990
	C	0.978	0.979	0.982	0.981	0.984	0.992	0.991	0.993	0.990	0.994
	D	0.985	0.989	0.990	0.992	0.991	0.989	0.992	0.996	0.993	0.994
	E	1.001	1.000	1.002	1.010	1.005	1.008	1.005	1.010	1.015	1.053
	F	0.996	0.999	1.003	1.002	1.003	1.010	1.005	1.015	1.045	1.060
	G	0.999	0.996	1.005	1.003	1.020	1.010	1.011	1.022	1.065	1.121
	H	0.999	1.002	1.004	1.005	1.009	1.015	1.019	1.030	1.089	1.156
	I	1.001	1.003	1.008	1.006	1.010	1.011	1.069	1.155	1.120	1.188
	J	1.010	1.011	1.009	1.010	1.011	1.023	1.088	1.145	1.165	1.190

[0075]

[Table 8]

Series resistance		First zinc oxide layer									
		A	B	C	D	E	F	G	H	I	J
Second zinc oxide layer	A	0.989	0.989	0.989	0.992	0.994	0.995	0.999	0.999	1.098	1.200
	B	0.990	0.991	0.994	0.999	0.999	1.001	1.000	1.002	1.090	1.201
	C	0.993	0.995	0.998	1.002	1.001	0.998	0.999	1.000	1.082	1.205
	D	0.996	0.996	0.999	0.997	0.989	0.992	1.001	1.003	1.102	1.197
	E	0.995	0.988	0.996	0.995	1.002	1.003	1.009	1.100	1.152	1.199
	F	0.993	0.991	0.997	1.020	1.001	1.005	1.012	1.120	1.135	1.140
	G	0.989	0.996	0.999	1.002	1.002	1.005	1.015	1.153	1.190	1.250
	H	0.989	0.999	0.999	1.011	1.010	1.010	1.015	1.160	1.203	1.203
	I	1.025	1.060	1.055	1.068	1.088	1.101	1.105	1.169	1.170	1.201
	J	1.150	1.155	1.102	1.100	1.111	1.056	1.177	1.188	1.165	1.250

[0076]

\*In the above-described relative series resistance, a larger numeral represents a higher series resistance. The electrical properties deteriorate as series resistance increases.

5 [0077]

[Example 2]

A stacked photovoltaic element was prepared in a manner similar to that for Example 1, by using a pin-type photovoltaic element 302 with the i-layer of  
10 intrinsic microcrystalline Si as the second photovoltaic element, a pin-type photovoltaic element 305 with the i-layer of intrinsic amorphous Si:H as the first photovoltaic element, and an intermediate layer of zinc oxide (refer to Fig. 3).

15 [0078]

The zinc oxide layer sample was prepared separately from the photovoltaic element on a quartz substrate, where the same Al-containing target was used, and charge rates of oxygen gas and vaporized H<sub>2</sub>O  
20 gas were adjusted to have a varying electric conductivity (Table 9). Based on these results, the conditions were found to prepare the zinc oxide layer having resistivity gradually changing in the thickness direction of the layer, so-called graded resistivity.

25 [0079]

The stacked photovoltaic element with the intermediate layer of zinc oxide was prepared in the

same manner as in Example 1, except that the zinc oxide layer was prepared under the conditions given in Table 10, where these conditions were adjusted to give zinc oxide resistivity increasing from the first photovoltaic element side towards the second photovoltaic element side.

[0080]

The stacked photovoltaic element samples thus prepared (Ex. 2-1 to 2-5) were evaluated in the same manner as in Example 1. The results are also given in Table 10.

[0081]

[Table 9]

Sample No.	Resistivity ( $\Omega\text{cm}$ )
A	$5.0 \times 10^{-1}$
B	$5.0 \times 10^0$
C	$2.5 \times 10^1$
D	$5.5 \times 10^1$
E	$5.5 \times 10^3$
F	$5.0 \times 10^3$
G	$7.5 \times 10^3$

[0082]

[Table 10]

Element No.	Resistivity range	Conversion efficiency
Ex. 2-1	$5.0 \times 10^0 - 5.0 \times 10^3$	1.000
Ex. 2-2	$5.0 \times 10^2 - 5.0 \times 10^3$	1.010
Ex. 2-3	$5.0 \times 10^0 - 4.0 \times 10^2$	0.979
Ex. 2-4	$5.0 \times 10^{-1} - 1.0 \times 10^3$	0.985
Ex. 2-5	$5.0 \times 10^0 - 7.5 \times 10^3$	0.987

[0083]

It is found also in Example 2 that the zinc oxide layer having a higher resistivity on the second photovoltaic element side and a lower resistivity on the first photovoltaic element side can increase reflection, improve short-circuit current and improve the joint surface of the stacked element, to improve its FF level, photocurrent and conversion efficiency, even when resistivity increases in a graded manner.

[0084]

It is also found that the effective resistivity range is  $2 \times 10^0 \Omega\text{cm}$  or more and  $5 \times 10^3 \Omega\text{cm}$  or less, and high resistivity range is  $5 \times 10^2 \Omega\text{cm}$  or more.

[0085]

[Effect of the Invention]

As explained so far, the present invention can provide a stacked photovoltaic element exhibiting a high conversion efficiency realized by efficiently absorbing light over an entire wavelength range of

incident light, reducing short-circuit current flowing  
an electrical defect and improving juncture between the  
zinc oxide layer and semiconductor layer.

[Brief Description of the Drawings]

5           [Fig. 1] A schematic cross-sectional view showing  
the structure of a conventional photovoltaic element  
provided with a zinc oxide layer.

          [Fig. 2] A schematic cross-sectional view showing  
the electrical polarities of a part of one embodiment  
10 of the stacked photovoltaic element of the present  
invention.

          [Fig. 3] A schematic cross-sectional view showing  
the structure of one embodiment of the stacked  
photovoltaic element of the present invention.

15          [Fig. 4] A schematic cross-sectional view showing  
the power-generating operation of the stacked  
photovoltaic element of the present invention.

          [Fig. 5] A schematic view illustrating one  
embodiment of a suitable apparatus for producing a  
20 semiconductor layer for the stacked photovoltaic  
element of the present invention.

          [Fig. 6] A schematic view illustrating one  
embodiment of a suitable apparatus for producing a zinc  
oxide layer for the stacked photovoltaic element of the  
25 present invention.

[Description of Reference Numerals or Symbols]

100 Stacked photovoltaic element



- 101 Substrate
- 102 Second photovoltaic element
- 103 Zinc oxide layer
- 104 First photovoltaic element
- 5 105 Transparent electrode
- 106 Short circuit in the second photovoltaic element
- 107 Short circuit in the first photovoltaic element
- 200 Stacked photovoltaic element of the present invention
- 10 201 Substrate
- 202 Second zinc oxide layer showing n- type electrical properties
- 203 First zinc oxide layer showing n+ type electrical properties
- 15 204 n-type semiconductor of first photovoltaic element showing n++ type electrical properties
- 205 Transparent electrode
- 206 Second photovoltaic element
- 207 Zinc oxide layer
- 20 208 First photovoltaic element
- 300 Stacked photovoltaic element of the present invention
- 301 Substrate
- 302 Second photovoltaic element
- 25 303 Second zinc oxide layer
- 304 First zinc oxide layer
- 305 First photovoltaic element

- 306 Transparent electrode
- 401, 403 Short circuit in the second photovoltaic element
- 402 Short circuit in the first photovoltaic element
- 5 500 Semiconductor deposited film forming apparatus
- 501 Load chamber
- 502 Chamber for n-layer
- 503 Chamber for i-layer of microcrystalline silicon
- 504 Chamber for i-layer of amorphous silicon
- 10 505 Chamber for p-layer
- 506 Unload chamber
- 507, 508, 509, 510, 511 Gate valve
- 512 Heater for heating microcrystal i-layer of substrate
- 15 513 Plasma CVD chamber for microcrystal i-layer
- 514 Heater for heating n-layer of substrate
- 515 Plasma CVD chamber for n-type layer
- 516 Heater for heating i-layer of amorphous silicon of substrate
- 20 517 Plasma CVD chamber for i-layer
- 518 Heater for heating p-layer of substrate
- 519 Plasma CVD chamber for p-layer
- 520 Holder transporting rail
- 600 Sputtering chamber
- 25 601 Substrate holder
- 602 Gas inlet tube
- 603 Heater

604 Zinc oxide target

605 DC power source

606 Earth shield

[Name of the Document] Abstract

[Abstract]

[Problem(s)] It is to provide a stacked photovoltaic element exhibiting a high conversion efficiency

5 realized by producing large photocurrent without causing decreased electromotive force.

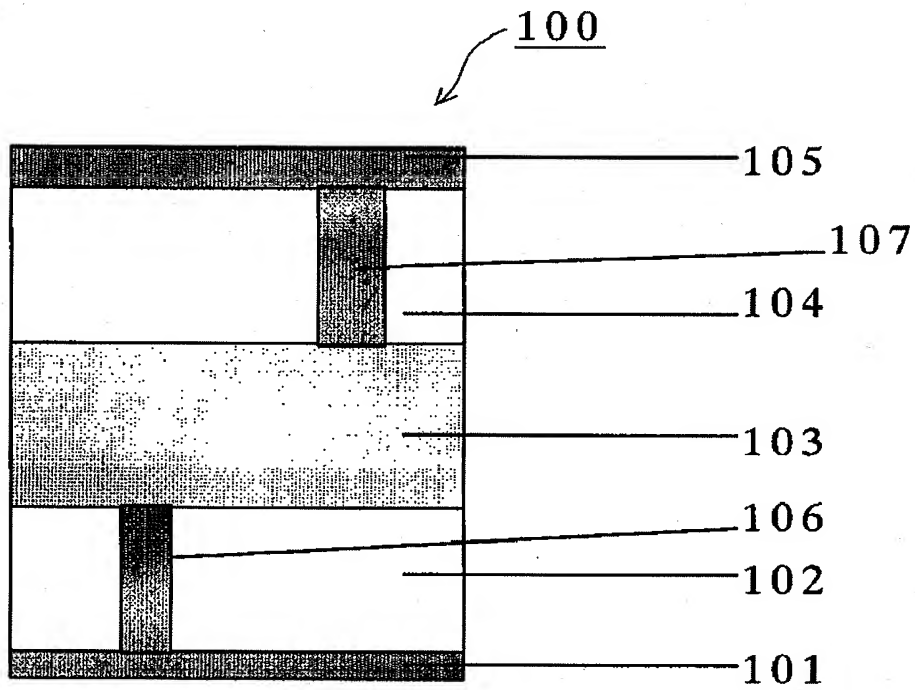
[Means for Solving the Problem(s)] A stacked photovoltaic element comprising a plurality of unit photovoltaic elements each composed of a pn- or pin-

10 junction, connected to each other in series, characterized in that a zinc oxide layer is provided at least one position between the unit photovoltaic elements, and the zinc oxide layer has resistivity varying in the thickness direction.

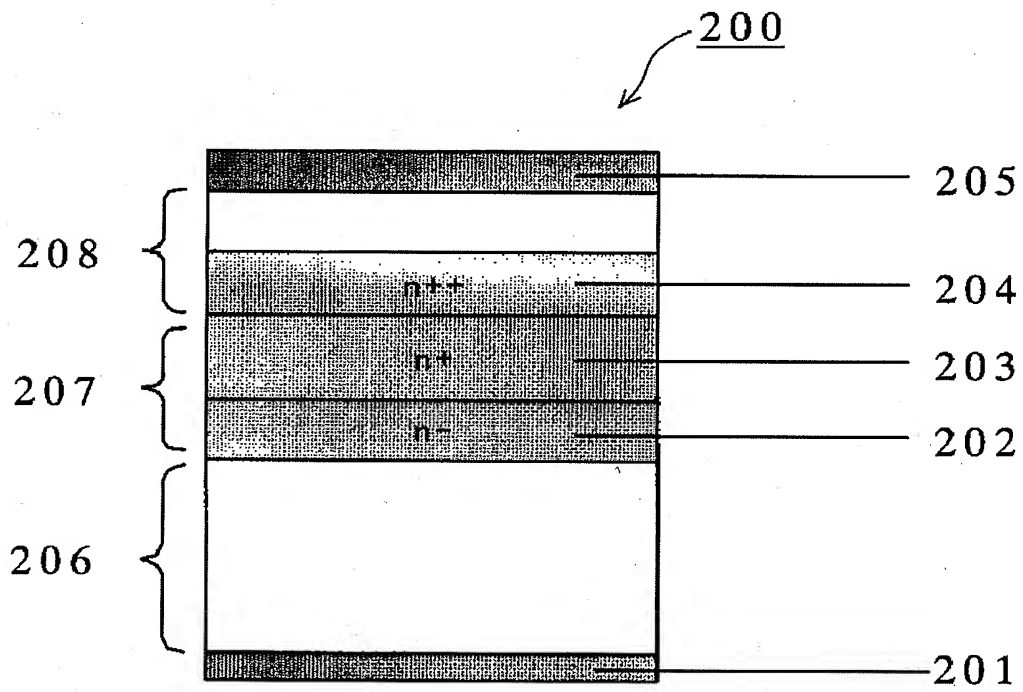
15 [Elected Drawing] Fig. 3

【書類名】 図面 [Name of Documents] Drawings

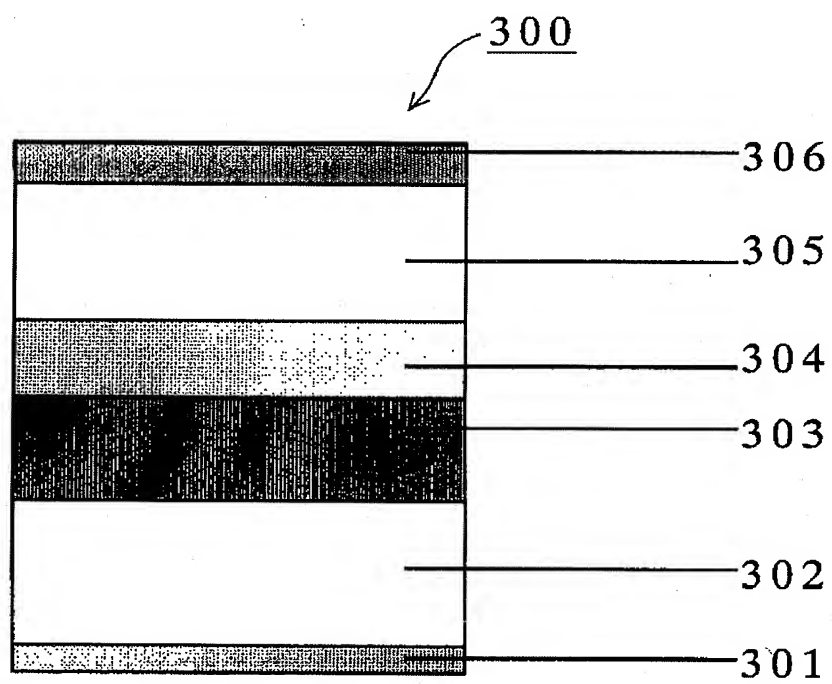
【図1】 Fig. 1



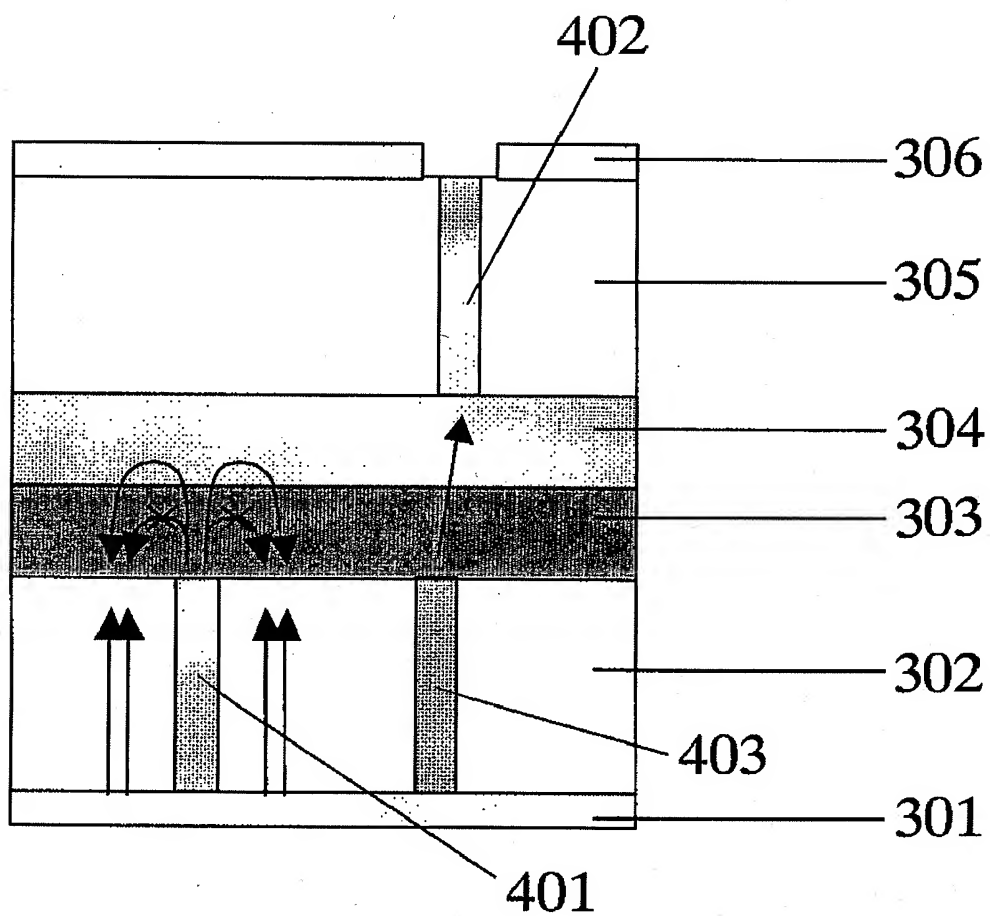
【図2】 Fig. 2



【図3】 Fig. 3

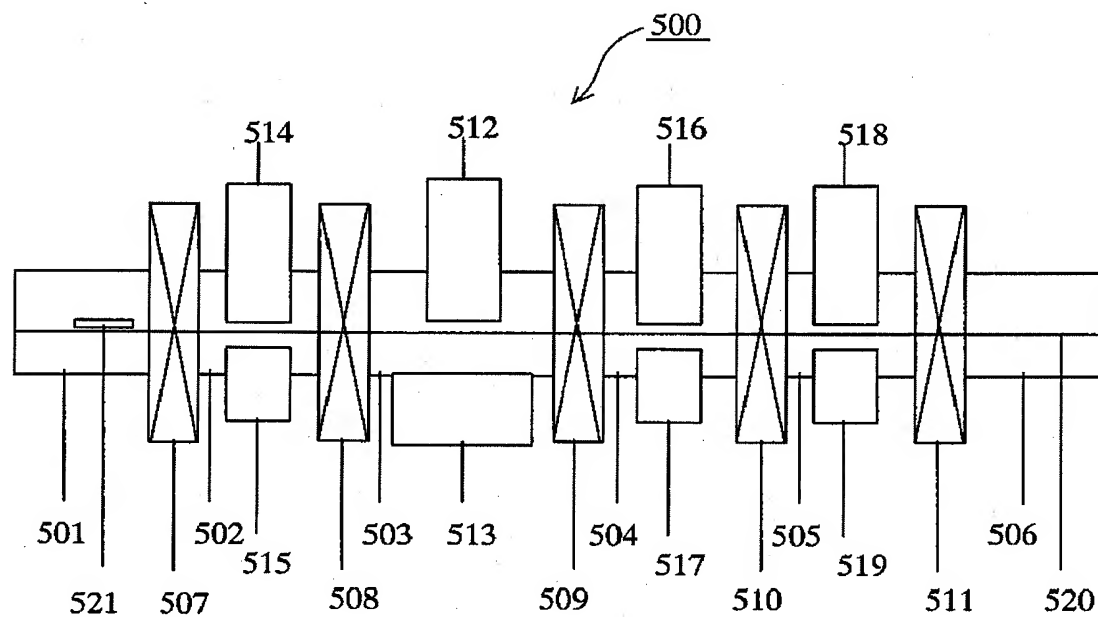


【図4】 Fig. 4





【図 5】 Fig. 5



【図6】 Fig. 6

